Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2018-1155-AC2, 2019

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Analysis of Sulfate Aerosols over Austria: A Case Study" *by* Camelia Talianu and Petra Seibert

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Received and published: 25 March 2019

Dear referee,

Thank you very much for the comments to our paper.

Here are the answers to your comments. In the following: "RefC" is the comment from Referee, "AuthR" is the author's response and "AuthCM" represents the author's changes to the manuscript. Page and line number refer to the page and line number in the version submitted for discussion.

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Discussion paper

Specific Comments

Comment 1.

RefC: Page 2, Lines 1-2 (Introduction): "worldwide in situ observations of refractory PM1 chemical composition have shown that the sulfate contribution may reach more than 50% of aerosol mass, depending on the location. See, for example, Zhang et al., 2007."

AuthR: This reference was added to the text and it was included in References list.

AuthCM: Page 2, Line 2: Added
"; worldwide in situ observations of refractory PM_1 chemical composition have shown that the sulfate contribution may reach more than 50% of aerosol mass, depending on the location (Zhang et al., 2007)."
Added reference (Zhang et al., 2007).

Comment 2.

RefC: Page 2, Lines 11-14 (Introduction): "a recent and important reference on SO2 sources worldwide, and also on sulfate radiative effects, is Yang et al., 2017."

AuthR: Added sentence to the text, referencing also the paper.

AuthCM: Added on Page 2, Line 14: “A recent review of SO_2 sources worldwide can be found in (Yang et al., 2017).”

Comment 3.

RefC: Page 2, Lines 18-20 (Introduction): "I recommend that you add a phrase or two to improve the description of the sulfate radiative effects, both direct and indirect. Also, you must include some key references for that."

AuthR: Done, see changes below.

AuthCM: Page 2, Line 18: added references AEROCOM project, IPCC AR5 for cooling effects of sulfate aerosol
Page 2, Line 20: Added text:
“The direct radiative effects are strongly correlated to the emission sources, while the indirect effects are correlated to both emission sources and cloud cover (Déandreis et al., 2012) (Yang et al., 2017).”

Comment 4.

RefC: Page 2, Lines 27-29 (Introduction): "Do you know of previous studies that promoted integration of data from in situ observations, remote sensing measurements and atmospheric transport modelling? I recommend that you provide an outlook of what has been done before, concerning to data integration from different platforms."

AuthR: In the last decade, the synergy of the in situ, remote sensing data and models was used in more atmospheric studies related to long-range transported aerosols and estimation of their potential sources (see for example A. Pappayanis et al. (Sci Total Environ. 2014;500-501:277-94. doi:10.1016/j.scitotenv.2014.08.101, 2014 - C.T. coauthor), D. Nicolae et al. (2013 - C.T. coauthor, Ansmann et al 2018, Eckhardt et al 2008 - P.S. coauthor, Cazacu et al 2012 - C.T. coauthor], [Sauvage et al 2017], [Chalbot et

al 2017],[D.G. Kaskaoutis et al., 2012]).

However, to our best knowledge, there have been no studies combining CAMS-based aerosol data with remote sensing and in situ measurements and transport models.

AuthCM: Added on Page 3, at the end of section “Introduction”:
“The synergy of the in situ, remote sensing data and models was used in more atmospheric studies related to long-range transported aerosols and estimation of their potential sources; see for example (Papayannis et al., 2014) for dust, (Nicolae et al., 2013) and (Ansmann et al., 2018) for fires, (Eckhardt et al., 2008) and (Cazacu et al., 2012) for volcanic ash, (Sauvage et al., 2017), (Chalbot et al., 2013) and (Kaskaoutis et al., 2012) for anthropogenic aerosols. However, to our best knowledge, there have been no studies combining CAMS-based aerosol data with remote sensing, in situ measurements and transport models. The assimilation of ground-based remote sensing measurements in CAMS is a long-term goal.”

Comment 5.

RefC: Page 2, Line 29 (Introduction): "please include a reference for NATALI aerosol typing model."

AuthR: Added reference for NATALI aerosol typing.

AuthCM: Page 2, Line 29 "... and NATALI aerosol-typing model, and atmospheric transport modeling." is replaced by "... and NATALI aerosol-typing model (Nicolae et al., 2018), and atmospheric transport modeling."

Comment 6.

RefC: Page 3, Line 15 (Methods): "You must give more details about the ground based air quality monitoring site and surroundings. Are there local air pollution sources affecting the site? How is the topography of the surroundings? What are the typical aspects of atmospheric circulation? Are there other air quality monitoring stations nearby?"

AuthR:

AuthCM: Added text to Page 3, Line 16
“Pillersdorf (315 m) is located in hilly terrain in the northeastern part of Austria, around 60 km north from Vienna. The station is a part of the national background monitoring network and an EMEP background monitoring station. The surroundings are mostly forests and agricultural areas far from strong anthropogenic sources. Austria belongs to the midlatitude climate belt, in the transition between maritime and continental climate, and the weather is dominated mostly by travelling highs and lows. The station provides:”

Comment 7.

RefC: Page 3, Line 15 (Methods): "I suggest that you include a description of the general aspects of climate and atmospheric synoptic scale circulation for the study region and season."

AuthR: The description was added to the text for Comment 6.

AuthCM: none

Comment 8.

RefC: Page 3, Line 25-28 (Methods): "It is very important to include a map showing the location of all stations explored in this manuscript. That will improve understandability for the readers that are not familiar with EARLINET and with general aspects of Europe geography."

AuthR: OK.

AuthCM: A map has been added to the Supplement.

Comment 9.

RefC: Page 5, Line 2 (Methods): "I suggest that you briefly explain (2-3 phrases) how a source-receptor model works. What do you need as input? Are there iterations required to tune the model parameters, in order to match model results and observations?"

AuthR: A more detailed explanation has been introduced. FLEXPART is not tuned or iterated.

AuthCM: Page 5 Line 2: Added after "...or gridded sources"
"(Seibert and Frank, 2004). The model ingests ECMWF 3D meteorological fields and solves the equations for transport, turbulent diffusions and other relevant processes in a Lagrangian framework (Stohl et al., 1998) (Pisso et al., 2019). The sensitivity of a receptor concentration to potential sources is obtained directly as the model output in the case of a backward run (Seibert and Frank, 2004) (Eckhardt et al., 2017)."

Comment 10.

RefC: Page 5, Line 12-14 (Methods): "the term “pure aerosol” usually refers to homogeneous particles made of a single chemical compound. This is not the case of aerosol classes like “continental”. Please find another term."

AuthR: Changed "pure aerosol" to "typical aerosol".

AuthCM: All occurrences of “pure aerosol” changed to “typical aerosol”.

Comment 11.

RefC: Page 5, Lines 28-32 (Methods): "it seems that there is a circular reasoning here: you aim to determine aerosol types from Lidar observations (Tables 3 and 6), but, at the same time, you have to assume aerosol types based on NATALI to make use of part of the Lidar observations. Please comment on that."

AuthR: The classes of typical aerosols in NATALI are defined based on the optical properties. If one of the properties is not measured, the type of the aerosol could still be identified based on the other measured properties, which are usually enough to constrain (approximately) the aerosol type. So, on Page 5, Line 28-32, the aerosol type is identified using particle depolarization ratio and AOD at different wavelengths. The lidar ratio is not measured, but it is assumed to be in the range attributed to that class. Please note that the validation of the NATALI model was performed on measurements having all properties measured. So, it is not a circular reasoning, but an estimation based on fully-characterized cases. There is no other way if the measurement is missing. As a cross-check, the type of the aerosol is constrained identifying the potential source with the transport model.

AuthCM: none

Comment 12.

RefC: Page 6, Lines 1-2 (Methods): "Please include a brief description (1-2 phrases) of the gradient method for detecting aerosol layers, and include more references for that. It is important to state the criteria used to identify an aerosol layer, to provide reproducibility of results. Also, please clarify that you applied the gradient method both for Lidar and CAMS profiles."

AuthR: Done, see changes to manuscript.

AuthCM: Page 6, Lines 1-2 changed to
"The aerosol layers are identified from the lidar measurements with the gradient method, applied to the RCS profiles (Belegante et al., 2014) (Nicolae et al., 2018). The gradient method is based on the identification of the peaks/valleys from the first derivative applied to the vertical profiles. If two consecutive layers are very close (less than 100 m), these layers are merged into one layer. Also, if the signal to noise ratio in the layer is lower than a threshold (here set to 5), the layer is discarded."
Page 6, Lines 24-26 changed to:
"The layers for the event at the in situ site are then determined by applying the same gradient method as for lidar data processing, but applied to the altitude profiles of aerosol concentrations. The concentrations are computed by multiplying the CAMS mixing ratios and the air density."

Comment 13a.

RefC: Page 6, Lines 11-15 (Methods): "the analysis of air quality timeseries were performed for how many years of data? How frequent were events like the one you described in the manuscript, in April 2014?"

AuthR: The analysis of air quality time series were performed for the spring and summer periods for the years 2010–2014. Events as described in the manuscript occur typically 1–2 times per year, between March and April. Unfortunately there were no lidar data to analyze the other events (no trajectories over lidar stations, no high-enough quality data for the corresponding periods). April 2010 was also dominated by the Eyjafjallajökull volcanic eruption.

AuthCM: none

Comment 13b.

RefC: Page 6, Lines 11-15 (Methods): "What is the objective criteria for “significant excess”?"

AuthR: The criterion for "significant excess" is 50% above the averaged values for 30 days.

AuthCM: Page 6, Line 13: changed "...is identified, " to "...is identified (values exceed by 50% the averaged values for 30 days),"

Comment 14.

RefC: Page 7, Line 14 (Methods): ""“The release is set to the location of the in situ station”. The word “release” is confusing in this context, because it gives the impression that pollutants were set to be released at the in situ station. Maybe “target” would work better here.""

AuthR: "release" was changed with "receptor".

AuthCM: "The receptor is set to the location"

Comment 15.

RefC: Page 7, Line 17 (Methods): "did you also consider SO2 biogenic sources, like oxidation from DMS? If not, how does it influence your results?"

AuthR: yes, the Flexpart model considers all the sulfate sources, including SO_2 biogenic sources. Also, for the trajectories at high altitude over ocean, the influence of biogenic sources is small.

AuthCM: none

Comment 16.

RefC: Figure 1: "SO2 lifetime in the troposphere is typically in the order of hours. Therefore, the SO2 observed at the ground based station may have a contribution of local sources, and possibly cannot be attributed to the regional transport (1-2 days) described in the case study. Please comment on that."

AuthR: As we mentioned before (see answer to comment 6), the station Pillersdorf is a regional background site, with no significant local anthropogenic sources, especially of SO_2 (by the requirements of EMEP "background site" and also because the SO_2 emissions are generally very low in Austria). The SO_2 measured at Pillersdorf is the result of the sulfate transported together with dust.

AuthCM: none

Comment 17.

RefC: Figures 2 and 3: "could you convert “model levels” to altitude, to improve understandability of the plots?"

AuthR: we would prefer to keep the model levels in these plots because the value (as altitude) of each model level is variable, it depends on the meteorological conditions (temperature, humidity, etc) that are variable in time. For each case, the altitude can be computed using the geopotential heights.

AuthCM: none

Comment 18.

RefC: Page 8, Lines 15-17 (Results): "if the diurnal evolution of sulfate and dust layers are correlated with SO2 and PM2.5, and dust is correlated with PM10, can I conclude that all variables are correlated? It would be interesting to see the diurnal evolution of ground based measurements and CAMS. In addition, is this correlation between CAMS and ground based measurements valid for all layers, or just for the lowermost layer?"

AuthR: There is a correlation between sulfate and dust layers and values of SO_2, $PM_{2.5}$ and PM_{10} from in-situ measurements, but it is important to analyze each case separately to find the correlation factor between them. Also, the correlation between CAMS and ground based measurements is valid for the lowermost layer (in situ data are assimilated in CAMS). For the rest of the layers, not much can be said as CAMS does not assimilate yet ground-based lidar data.

AuthCM: none

Comment 19.

RefC: Figure 4: "the legend is illegible, text must be enlarged. To better interpret this Figure, it is important to know which profiles correspond to daytime and nighttime (i.e., local time of each plot). Information on the typical planetary boundary layer height at Pillersdorf would also help. It would be interesting to point out whether and when there was an input of aerosols from upper layers to the boundary layer, affecting air quality at Pillersdorf."

AuthR: We have improved the figure and added a sentence on boundary-layer heights. We also added in the caption of Fig. 4 the UTC–local time difference.

AuthCM: Changed fonts in the legend of Fig. 4; modified caption: changed “aerosol” to “total aerosol”; added “Local time is UTC+2.”.

Added on Page 6, Line 31:
“During the period under investigation, with low wind speeds and mostly clear skies, the boundary-layer height varied at Pillersdorf from less than 100 m at night to about 1500 m in the afternoon.”

Comment 20.

RefC: Figure 5: "there are too many lines (altitudes) in the lower plots of sub-figures, it is difficult to interpret. There must be a compromise between completeness and understandability. I suggest that you keep only 3-4 representative altitudes (low, medium, high)."

AuthR: The figure was changed, keeping only the trajectories passing over the lidar stations involved in this analysis.

AuthCM: New Fig. 5

Comment 21.

RefC: "Figures 6, 10c, 11c, 12d: you must indicate the locations of the monitoring stations in the maps."

AuthR: The figures were changed adding the location of the monitoring stations.

AuthCM: New figures 6, 10c, 11c, 12d

Comment 22.

RefC: Figure 10c: "the model calculates SO2 $<$ 1 ug/m3 for layer 1, which is inside the boundary layer, all over Europe. How does it compare to your ground based measurements?"

AuthR: There was a bug in superposing the concentration distribution on map for the zoomed distribution (only). The figures 10c, 11c and 12d were replaced with figures with correct data.

There is a good agreement between model and ground based measurements, as can be seen from Fig. 1 and Fig. 10c.

AuthCM: the figures 10c, 11c and 12c were replaced.

Comment 23.

RefC: Table 3: "can you see changes on aerosol properties as they are transported? For example, layers 1 and 2 at Leipzig and Pillersdorf are associated (Table 1). How does aerosol intrinsic and extrinsic properties change along this $\approx$ 24h transport?"

AuthR: Yes, changes on aerosol properties can be observed for the correlated layers. For layer 1, the aerosol transported from Leipzig to Pillersdorf is mixed with the aerosols accumulated inside the PBL (the trajectory is under 1500 m), leading to an increase of the sulfate fraction in aerosols. The aerosol size increase also. These are reflected by the decrease in the lidar ratio and AE, measured at Leipzig and computed at Pillersdorf.

For Layer 2 (above PBL), the depolarization ratio, sulfate fraction and AE decreases, due to chemical processes (aging) and aerosol removal processes. The lidar ratio slightly increases, due to the decrease of the sulfate fraction from aerosol.

AuthCM: none

Comment 24.

RefC: Page 9, Lines 19-24: "since you did not discuss April 4 in detail, I recommend moving it to the supplementary material, as well as the corresponding figures and tables."

AuthR: We would prefer to keep the event in the article, as it is consistent with the event from Apr 02 and emphasize the main message of the paper (long-range transport of sulfate aerosols must be considered for local changes, as it can have non-negligible effects).

AuthCM: none

Comment 25.

RefC: Page 10, Lines 10-28: "the discussion of trajectories and source regions is rather qualitative. Terms like “medium to smaller contributions” are vague. Could you estimate percent contributions? Also, it is important to recognize limitations and uncertainties of the method."

AuthR: I could compute the contributions of the sources, but they would be obtained from the transport model, they would be a rough estimation, therefore I prefer to give only qualitative values. For confident quantitative results, I would need more measured data, time-dependent, not only monthly averaged sources, to be able to compute the central value and to compute the uncertainties.

AuthCM: none

Comment 26.

RefC: Page 10: "meteorological maps for the case study period would help to support the conclusions on aerosol transport. Particularly, trajectories calculated below 2000 m are more prone to uncertainties."

AuthR: The meteorological maps will be added to the Supplement of the paper.

AuthCM: meteorological maps in the supplement.

Comment 27.

RefC: Figure 16: "it does not contribute significantly to the discussion. I suggest to exclude this figure, or to move it to the supplementary material."

AuthR: OK

AuthCM: Fig. 16 was moved to the Supplement of the paper.

Comment 28.

RefC: Page 11 (conclusion): "what are the main advantages of the method you used for this case study, compared to previous database-integration studies? What are the main limitations? How can the method be improved?"

AuthR: I think this is covered by the text added in Introduction as answer to Comment 4 and by the text from Page 11, lines 25 - 31.

AuthCM: none

Technical corrections

Comment 29.

RefC: Page 2, Line 17: "what do you mean by “key properties”? Optical? Physical? Be more specific."

AuthR: "key properties" means "optical, physical and chemical properties".

AuthCM: Page 2, Line 17
"The key properties" was changed to "The optical, physical and chemical properties"

Comment 30.

RefC: Page 5, Line 11: "omit the word “Ref.” before citing a reference. That occurs all through the manuscript, please check."

AuthR: OK.

AuthCM: removed “Ref. “

Comment 31.

RefC: Page 6, Line 6: "“The values of the CAMS quantities”: please use a more specific term, instead of “quantities”."

AuthR: "the CAMS quantities” were specified in text at page 4, line 22 - line 28.

AuthCM: Page 6, Line 6: “... the CAMS quantities” was changed to “... the CAMS products (mixing ratios, temperature, specific humidity, etc)”

Comment 32.

RefC: Figure 7: "scales are illegible."

AuthCM: Figure 7 The fonts for scales were increased to be more legible.

Comment 33.

RefC: Figures 7 and 8: "could be merged into a single figure. I recommend reformulation of the lidar plots adopting a standard pattern for the contourplots."

AuthR: The lidar plots are taken in the format available in the Earlinet database. I prefer to keep the format used by the Earlinet database for consistency.

AuthCM: none

Comment 34.

RefC: Figure 9: "what are the units for the color map? In addition, the units of longitude should be “degrees”, and not “degrees E”."

AuthR: The units for color map are seconds. The units of longitude and latitude were corrected.

AuthCM: the figures 9, 10, 11, 12 and 15 were replaced with the figures with units.

Comment 35.

RefC: Table 3: "Please define abbreviations in the table caption, to facilitate interpretation."

AuthR: The abbreviations are defined in the text.

AuthCM: none

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-1155, 2018.

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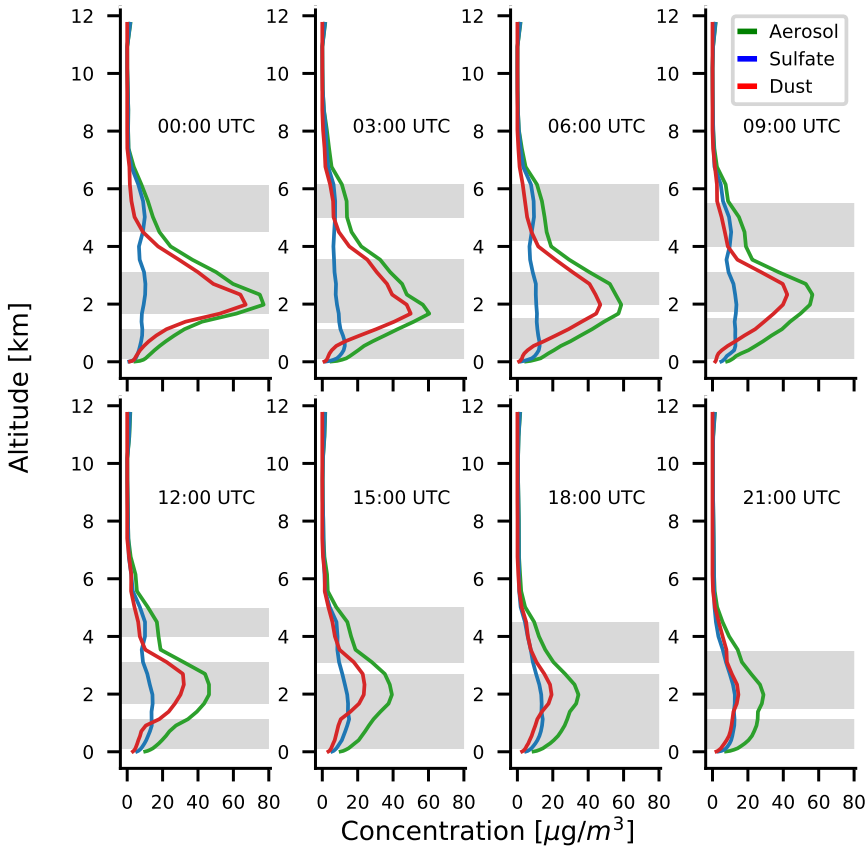

Fig. 1. CAMS total aerosol, sulfate and dust profiles for 02 April 2014, Pillersdorf. Grayed area represents the identified sulfate layers. Altitudes are given in km AGL. Local time is UTC+2.

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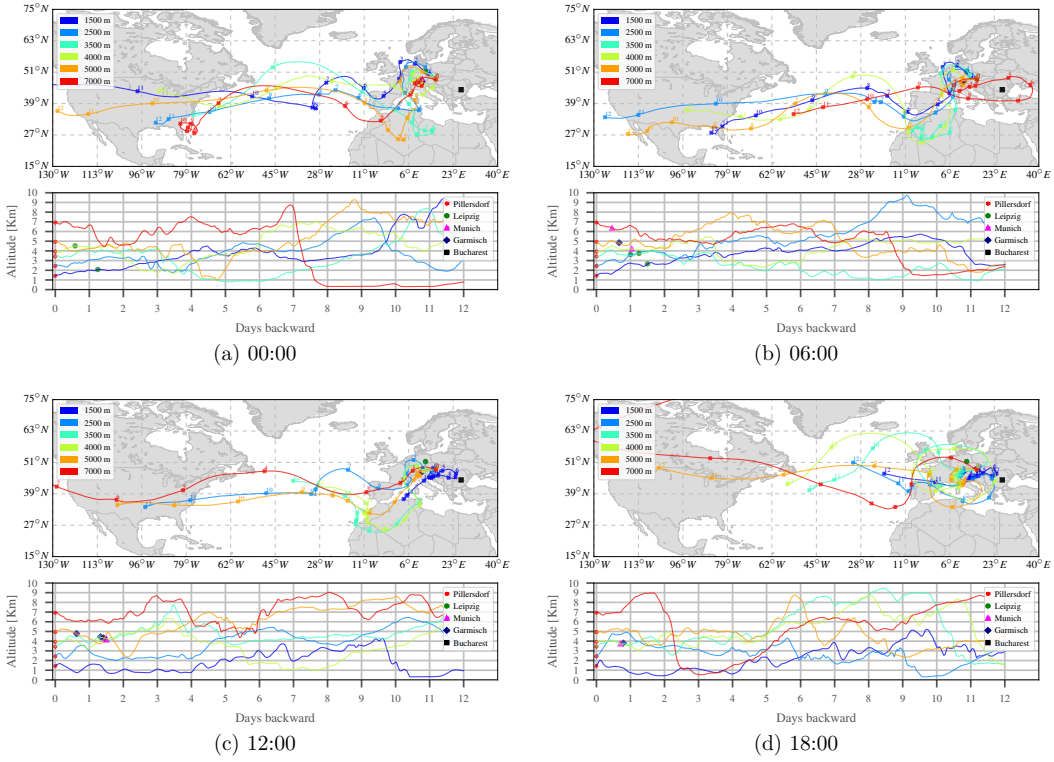

(a) 00:00 (b) 06:00

(c) 12:00 (d) 18:00

Fig. 2. Pattern of back-trajectories (upper plot of sub-figure) and their altitude profile, including overpassed lidar stations (lower plot of sub-figure) for Pillersdorf, 02 April 2014.

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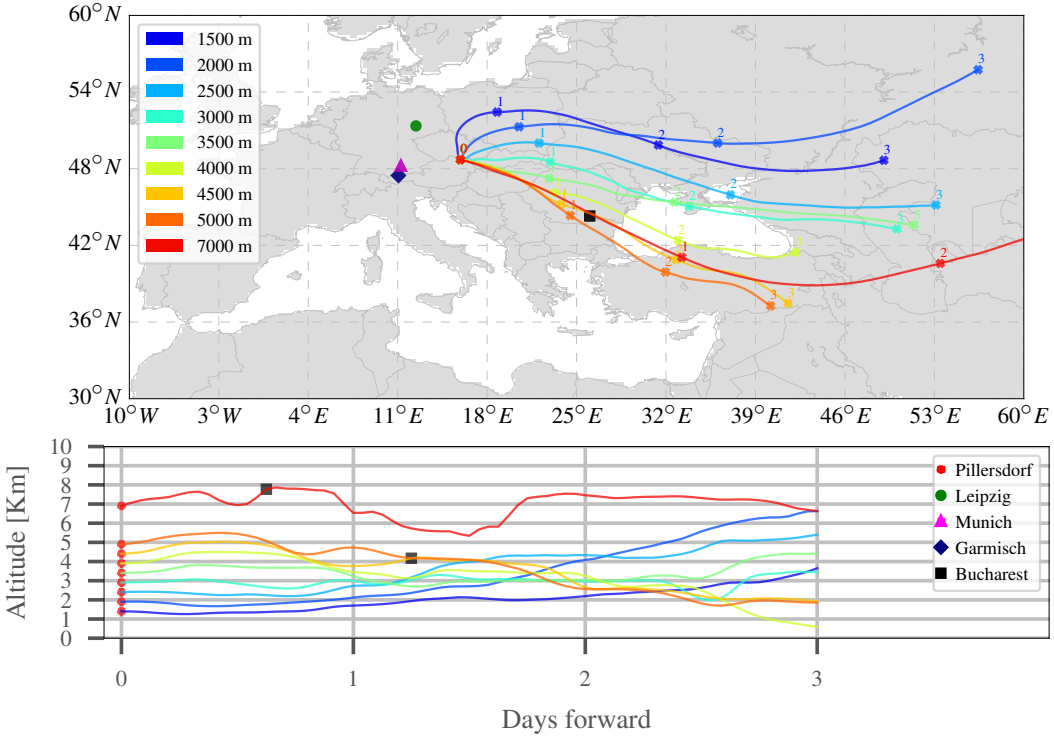

Fig. 3. Pattern of forward-trajectories (upper plot) and their altitude profile, including over-passed lidar stations (lower plot) for Pillersdorf, 02 April 2014, 06:00.

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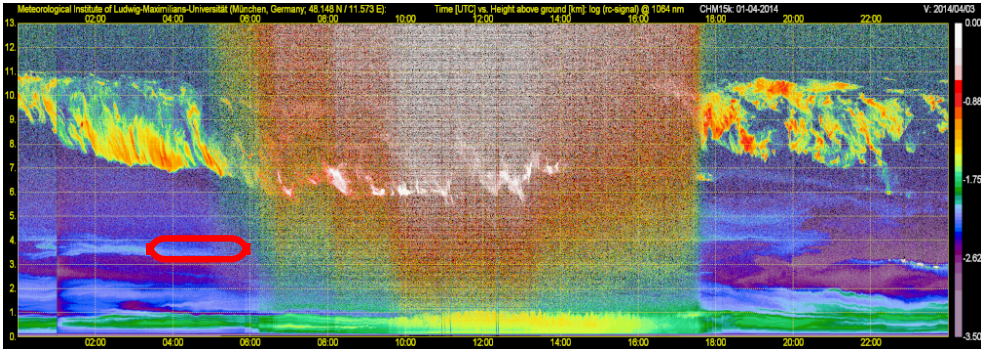
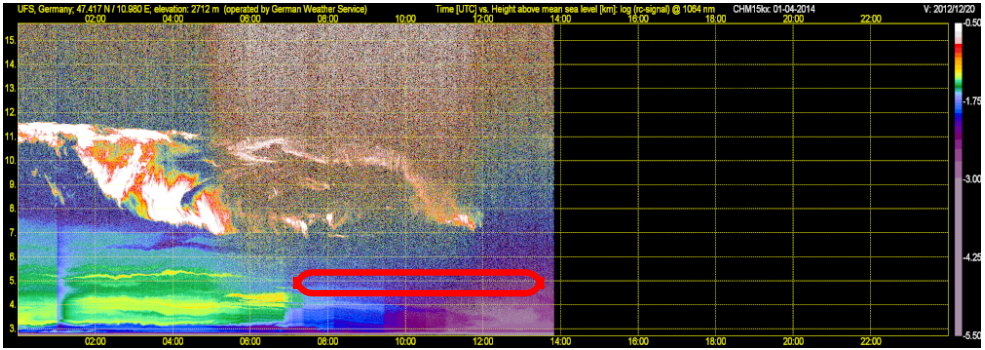


(a) Munich, 01 April 2014 Ceilometer YALIS



(b) Garmisch, 01 April 2014 Ceilometer

Fig. 4. Logarithm of the range corrected signal at 1064 nm, 24 h, for Munich (a) and Garmisch (b) stations. The red line boxes represent the identified layers.

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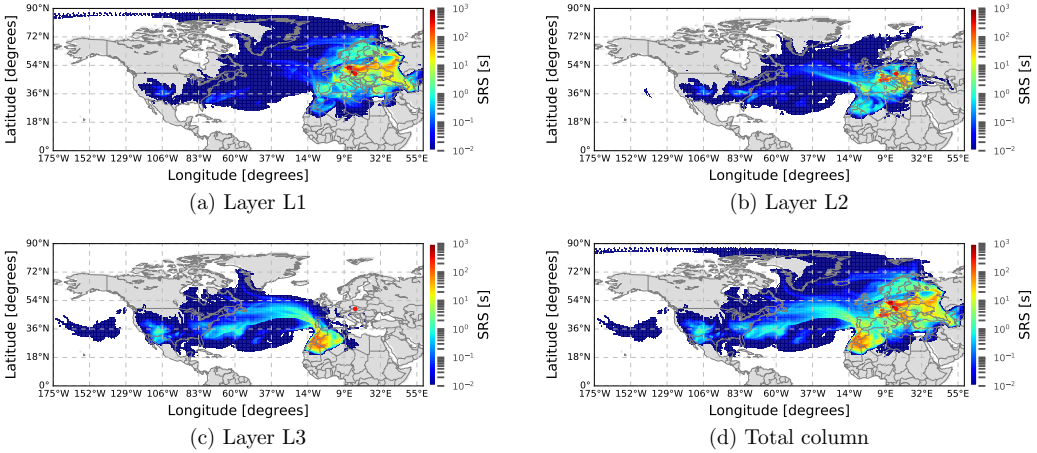

(a) Layer L1 (b) Layer L2

(c) Layer L3 (d) Total column

Fig. 5. Source-receptor sensitivity for layer L1 (a), L2 (b) and L3 (c) and total column (d), Pillersdorf, 02 April, 6:00.

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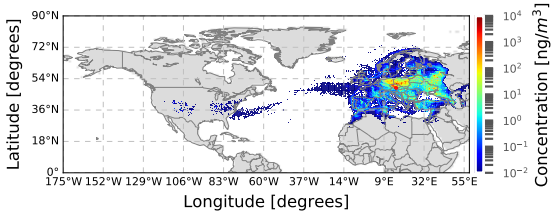
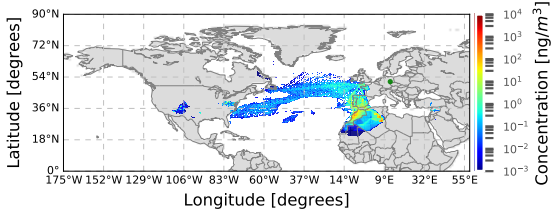
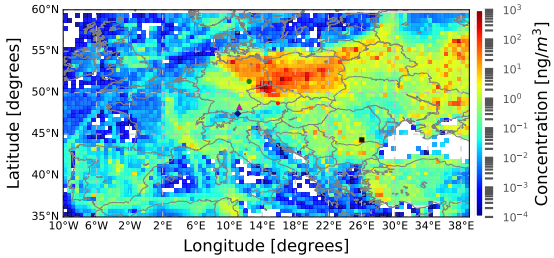


(a) Pillersdorf, 02 April, 06:00, layer L1

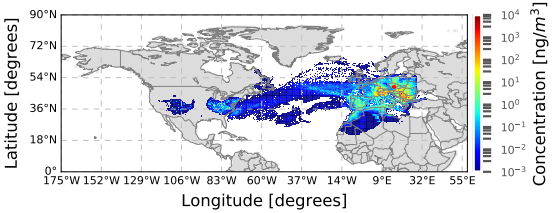


(b) Leipzig, 31 March, 18:00, layer corresponding to L1

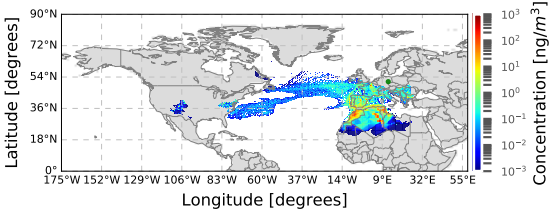


(c) Pillersdorf, 02 April, 06:00, layer L1, zoomed

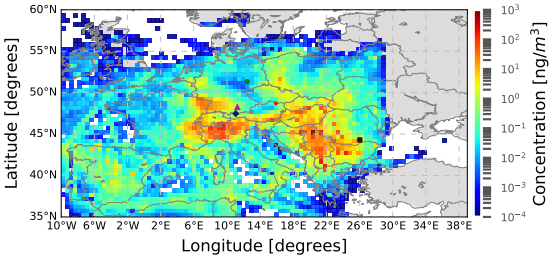
Fig. 6. Relative distributions of SO2 sources for Pillersdorf layer L1 (a), Leipzig (b); zoomed distribution for Pillersdorf layer L1 (c).



(a) Pillersdorf, 02 April, 06:00, layer L2

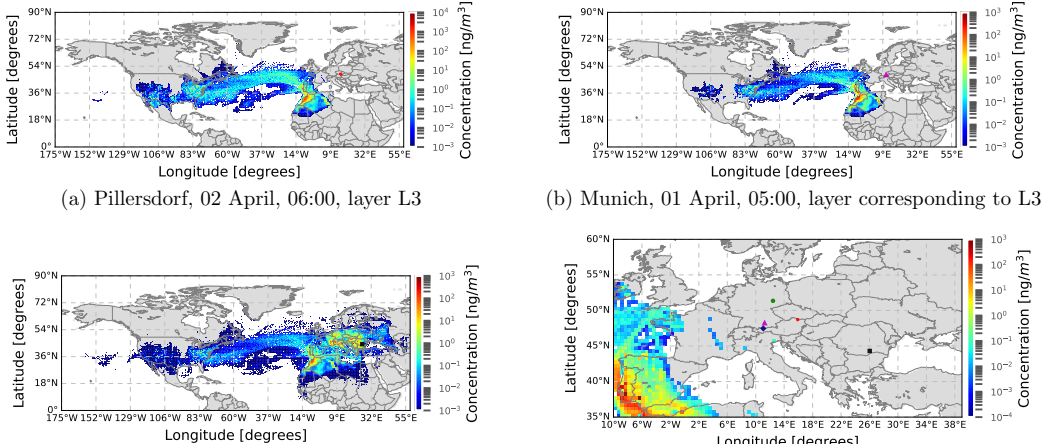


(b) Leipzig, 31 March, 23:00, layer corresponding to L2



(c) Pillersdorf, 02 April, 06:00, layer L2, zoomed

Fig. 7. Relative distributions of SO2 sources for Pillersdorf layer L2 (a), Leipzig (b); zoomed distribution for Pillersdorf layer L2 (c).



(a) Pillersdorf, 02 April, 06:00, layer L3

(b) Munich, 01 April, 05:00, layer corresponding to L3

(c) Bucharest, 03 April, 13:00, layer corresponding to L3

(d) Pillersdorf, 02 April, 06:00, layer L3, zoomed

Fig. 8. Relative distributions of SO2 sources for Pillersdorf layer L3 (a), Munich (b), Bucharest (c); zoomed distribution for Pillersdorf layer L3 (d).

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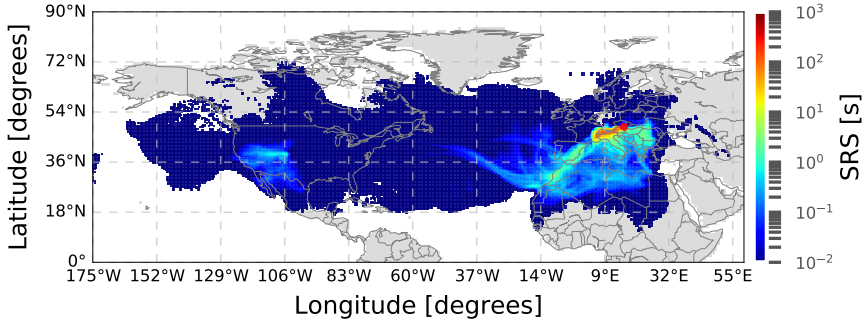
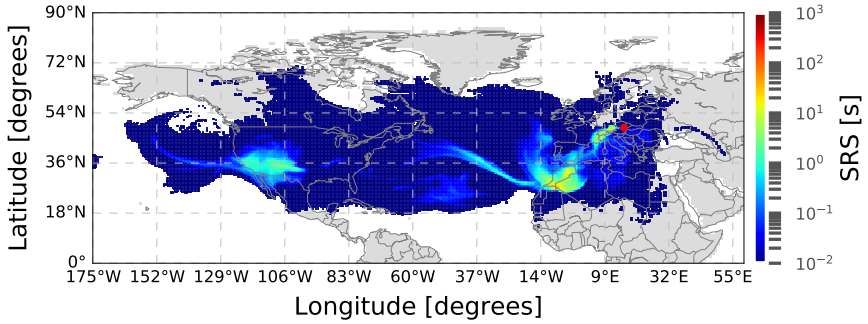


(a) Pillersdorf, 04 April, 12:00, layer L1



(b) Pillersdorf, 04 April, 12:00, layer L2

Fig. 9. Source-receptor sensitivity for layer L1 (a) and L2 (b), Pillersdorf, 04 April, 12:00

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